Symmetric photon-photon coupling by atoms with Zeeman-split sublevels

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We propose a simple scheme for highly efficient nonlinear interaction between two weak optical fields. The scheme is based on the attainment of electromagnetically induced transparency *simultaneously for both fields* via transitions between magnetically split $F=1$ atomic sublevels, in the presence of two driving fields. Thereby, equal slow group velocities and symmetric cross coupling of the weak fields over long distances are achieved. By simply tuning the fields, this scheme can either yield giant cross-phase modulation or ultrasensitive two-photon switching.

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The weakness of optical nonlinearities in conventional media precludes the effective interaction of extremely feeble fields containing few photons only [1]. This weakness sets a limit on the performance of ultrasensitive photonic elements (switches and couplers), as well as on nonclassical (“squeezing”) effects. It is also the main impediment towards constructing quantum logic gates, quantum teleportation, and cryptography schemes operating at the few-photon level [2]. The weakness of optical nonlinearities can be compensated by photon confinement in a high-Q cavity [3]. A promising avenue has been opened by studies of enhanced nonlinear coupling via electromagnetically induced transparency (EIT) in atomic vapors in the presence of classical driving fields, which induce coherence between atomic levels [4,5]. These studies have predicted the ability to achieve an appreciable nonlinear phase shift of extremely weak optical fields [6] or a two-photon switch [7,8], using the driven N-configuration of atomic levels. The main hindrance of such schemes is the *mismatch* between the group velocities of the field that is subject to EIT and its nearly free propagating partner, which severely limits their effective interaction length [9].

In the present paper we propose a scheme that can remove this bottleneck, by basically modifying the nonlinear interaction of weak optical fields: in contrast to all currently known schemes, it affects both fields in a completely *symmetric* fashion, rendering their group velocities equal. It thereby allows their cross coupling over very long distances and brings it to its ultimate limit of efficiency. This scheme relies solely on an *intra-atomic process* that causes simultaneous EIT for both fields interacting with magnetically (Zeeman-) split sublevels in the presence of two driving fields. Remarkably, by simply tuning the fields, this scheme can either yield giant cross-phase modulation or ultrasensitive two-photon switching in vapor [10]. It may, therefore, substantially advance the optical processing and communication of quantum information in conventional media, without resorting to photonic crystals [11] or resonators [12].

We first outline the proposed setup. Two weak optical fields E_a and E_b, having σ polarization along the y axis, propagate in the atomic medium along the z axis. Two π-(x-)polarized continuous wave (cw) driving fields $E_{d_{1,2}}$ and a static x-oriented magnetic field B are applied (Fig. 1, left inset). In the absence of these fields, the lower (L) and upper (U) atomic levels, having equal total angular momenta $F_L = F_U = 1$ (e.g,, the $D1$ line in ^{23}Na or ^{87}Rb), are sepa-

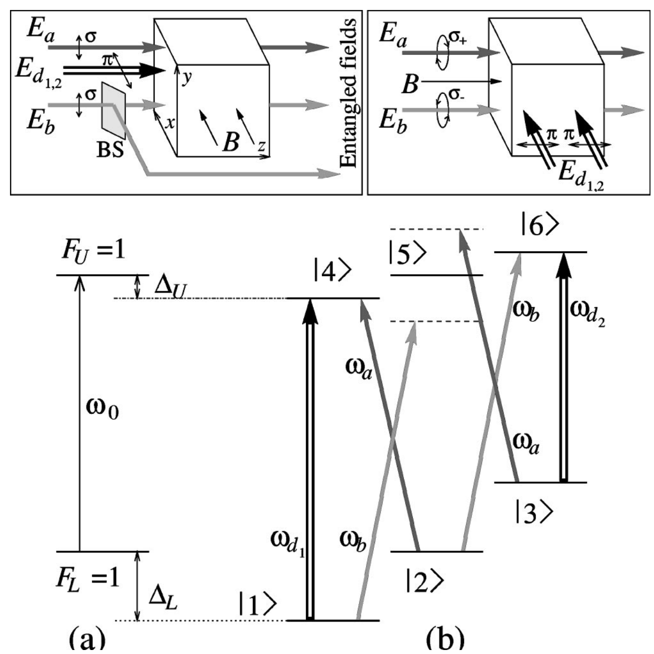

FIG. 1. Left inset: copropagating weak σ-polarized optical fields $E_{a,b}$, and strong π-polarized driving fields $E_{d_{1,2}}$ pass through the atomic vapor cell that is placed in a transverse magnetic field B. The beam splitter (BS) splits the E_b field into two components, one of which passes through the cell and interacts with the E_a field. The E_a and E_b fields at the output are then entangled. Right inset: perpendicular arrangement of the weak and driving fields suitable for a cold atomic gas. (a) Two degenerate atomic levels with angular momenta $F_L = F_U = 1$ are separated by the unperturbed energy difference $\hbar\omega_0$. (b) Magnetic field B splits each level into three Zeeman components with relative shifts Δ_L and Δ_U. Two Λ systems, $\Lambda_a \equiv |2\rangle \leftrightarrow |4\rangle \leftrightarrow |1\rangle$ and $\Lambda_b \equiv |2\rangle \leftrightarrow |6\rangle \leftrightarrow |3\rangle$, sharing ground state $|2\rangle$, are formed in the presence of $E_{d_{1,2}}$ ($\omega_{d_{1,2}}$) and give rise to EIT for the $E_a(\omega_a)$ and $E_b(\omega_b)$ fields, respectively. Transitions $|1\rangle \rightarrow |5\rangle$ and $|3\rangle \rightarrow |5\rangle$ serve to cross couple Λ_a and Λ_b with the ω_b and ω_a photons, respectively.

*URL: http://www.weizmann.ac.il/chemphys/gershon/; electronic address: gershon.kurizki@weizmann.ac.il rated by the frequency ω_0 [Fig. 1(a)]. The magnetic field B splits the lower and upper triplets of the atom into three components each, labeled $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$, $|5\rangle$, $|6\rangle$, respectively [Fig. 1(b)]. By appropriately tuning the fields, as detailed below, two symmetrically cross-coupled Λ configurations, $\Lambda_a \equiv |2\rangle \leftrightarrow |4\rangle \leftrightarrow |1\rangle$ and $\Lambda_b \equiv |2\rangle \leftrightarrow |6\rangle \leftrightarrow |3\rangle$, are realized and give rise to EIT for the two weak fields simultaneously.

Let us specify the transitions and the fields involved. The Zeeman shift of the sublevels in the lower and upper level is given by $\Delta_{L,U} = (\mu_B/\hbar) M_{L,U} g_{L,U} B$, where μ_B is the Bohr magneton, $g_{L,U}$ is the gyromagnetic factor of the corresponding atomic level, and $M_{L,U}$ is the magnetic quantum number of the corresponding state. The σ-polarized E_a and E_b fields act only on the transitions with $\Delta M = \pm 1$, while the π-polarized $E_{d_{1,2}}$ fields couple the states with $\Delta M = 0$ ($M = 0$). The frequencies of the cw driving fields $E_{d_{1,2}}$ are resonant with the atomic transitions $|1\rangle \to |4\rangle$ and $|3\rangle \to |6\rangle$, respectively, $\omega_{d_{1,2}} = \omega_0 \pm \Delta_D$, where $\Delta_D = \Delta_L - \Delta_U$. The E_{d_1} and E_{d_2} fields, having the same Rabi frequency Ω_d, act also on the transitions $|3\rangle \to |6\rangle$ and $|1\rangle \to |4\rangle$, with the detunings $\mp 2\Delta_D$, respectively. If $\Delta_D \gg |\Omega_d|$, this off-resonant coupling merely induces the ac Stark shifts $\mp |\Omega_d|^2/2\Delta_D$ of the states $|1\rangle$ and $|3\rangle$, which can be incorporated into the energy of the atomic state. This reasoning is valid, provided the detuning Δ_D is larger than the Doppler width of the atomic resonance $k\overline{v}$, where $k = \omega_0/c$ and $\overline{v} = \sqrt{3k_B T/m}$ is the mean thermal velocity of the atoms. The frequencies of the weak fields $E_{a,b}$ are chosen to be at Raman resonance with the corresponding two-photon transitions $|2\rangle \to |1\rangle$ and $|2\rangle \to |3\rangle$: $\omega_{a,b} = \omega_0 \mp \Delta_U + \delta_{a,b}$, where $\delta_{a,b}$ denotes a small detuning from the corresponding two-photon transition.

The cross coupling of the weak fields is achieved via the E_b-induced transition $|1\rangle \to |5\rangle$ and the E_a-induced transition $|3\rangle \to |5\rangle$. The fields are detuned from the corresponding transitions by the amounts $\pm \Delta_D$. The realization of the *dispersive cross coupling* requires that $\Delta_D \gg |\Omega_{a,b}|$, where $\Omega_{a,b}$ is the Rabi frequency of the corresponding field. The undesirable atom-field couplings via the transitions $|1\rangle \to |5\rangle$ and $|2\rangle \to |6\rangle$ for E_a and via the transitions $|3\rangle \to |5\rangle$ and $|2\rangle \to |4\rangle$ for E_b, can then be neglected, due to the large detunings $\pm(\Delta_L + \Delta_U)$ and $\pm 2\Delta_U$, respectively.

In an alternative setup, one can get rid of these couplings using the (circularly) σ_+ polarized E_a and σ_- polarized E_b fields propagating along the magnetic field lines (Fig. 1, right inset). Then the E_a field will act only on the transitions with $\Delta M = +1$, that is, $|2\rangle \to |4\rangle$ and $|3\rangle \to |5\rangle$, while the E_b field will act on $\Delta M = -1$ transitions, that is $|2\rangle \to |6\rangle$ and $|1\rangle \to |5\rangle$. However, in order to cancel the Doppler broadening of the EIT resonances in this setup, one would have to work with cold atoms ($T \leqslant 0.5\ \mu$K). By contrast, in the collinear Doppler-free geometry of Fig. 1, left inset, the EIT resonances can be very sharp at considerably higher temperatures.

Under optimal conditions for cross coupling in both setups discussed above (Fig. 1, left and right insets), we obtain the following set of coupled equations for the slowly varying probability amplitudes A_j of the six atomic states:

$$\partial_t A_1 = i\delta_a A_1 - i\Omega_d^* A_4 + i\Omega_b^* e^{-i\Delta_D t} A_5, \tag{1a}$$

$$\partial_t A_2 = -i\Omega_a^* A_4 + i\Omega_b^* A_6, \tag{1b}$$

$$\partial_t A_3 = i\delta_b A_3 - i\Omega_a^* e^{i\Delta_D t} A_5 + i\Omega_d^* A_6, \tag{1c}$$

$$\partial_t A_4 = -[\gamma - i(\delta_a - kv)]A_4 - i\Omega_d A_1 - i\Omega_a A_2, \tag{1d}$$

$$\partial_t A_5 = -[\gamma - i(\delta_b + \delta_a - kv)]A_5 + i\Omega_b e^{i\Delta_D t} A_1 - i\Omega_a e^{-i\Delta_D t} A_3, \tag{1e}$$

$$\partial_t A_6 = -[\gamma - i(\delta_b - kv)]A_6 + i\Omega_b A_2 + i\Omega_d A_3, \tag{1f}$$

where γ is the relaxation rate of states $|4\rangle$, $|5\rangle$, and $|6\rangle$, which we include here *phenomenologically* [7,9]. In deriving these equations, we have replaced the wave numbers $k_j (j = a,b,d_1,d_2)$ by k and, consistently with the discussion above, have assumed that the ac Stark shifts of the states $|1\rangle$ and $|2\rangle$ due to the off-resonance interaction with the E_{d_2} and E_{d_1} fields are incorporated in Δ_D. Obviously, in the cold-atom setup (Fig. 1, right inset) the velocity of atoms v is zero. In Eqs. (1), the sign of each term containing a Rabi frequency is determined by the Clebsch-Gordan coefficient for the corresponding atomic transition.

We neglect the depletion of the cw driving fields, for reasons explained below. In the perturbative solution of Eqs. (1), we shall assume that the relaxation time of the excited atomic states is short compared to the duration τ of the pulses $E_{a,b}$, $\tau \gg \gamma^{-1}$. Therefore, the slowly varying envelope approximation is well justified, since the pulse amplitudes change very little during an optical cycle. Then the evolution of the two weak fields $E_j (j = a,b)$ is governed by the following propagation equations:

$$[\partial_z + v_g^{-1}\partial_t]E_j = i\alpha_j E_j, \tag{2}$$

yielding $E_j(z,t) = E_j(0, t - z/v_g)\exp(i\int_0^z \alpha_j dz)$. Here, the macroscopic complex polarizabilities α_j given by

$$\alpha_a = \frac{\alpha_0 \gamma}{\Omega_a}\langle A_2^* A_4 + A_3^* A_5 e^{i\Delta_D t}\rangle_T, \tag{3a}$$

$$\alpha_b = \frac{\alpha_0 \gamma}{\Omega_b}\langle A_2^* A_6 + A_1^* A_5 e^{-i\Delta_D t}\rangle_T, \tag{3b}$$

are proportional to the linear resonant absorption coefficient $\alpha_0 = |\mu|^2 \omega_0 N/(2\epsilon_0 c\hbar\gamma) \equiv \sigma_0 N$, where σ_0 is the resonant absorption cross section and N is the density of atoms. In Eqs. (3), the averaging $\langle \cdots \rangle_T$ is performed over the atomic thermal motion, which obeys the Maxwellian distribution $W(v) = (u\sqrt{\pi})^{-1}\exp(-v^2/u^2)$.

The crucial point about Eq. (2) is that, due to the *symmetry* of the system with respect to the two fields E_a and E_b, their group velocities $v_g = [1/c + \partial\, \mathrm{Re}(\alpha_j)/\partial\delta_j]^{-1}$ are *equal*, as can be verified from Eqs. (3) and (1).

We assume that initially the driving fields optically pump all the atoms into the energy state $|2\rangle$. In the weak-field limit (much less than one photon per atom), the Rabi frequencies of the two interacting fields satisfy the condition $|\Omega_{a,b}| \ll \gamma, |\Omega_d|$. Then, during propagation, nearly all of the atomic population remains in $|2\rangle$ $(A_2 \simeq 1)$. This justifies the neglect of the driving-field depletion (assumed above) after the optical pumping is completed. Under these conditions, Eqs. (1) can be solved using fourth-order perturbation theory. Then, at the Raman resonance for both fields, $\delta_a = \delta_b = 0$, the polarizabilities are given by

$$\alpha_{a,b} = \frac{2i\alpha_0\gamma|\Omega_{b,a}|^2}{(\gamma \pm i\Delta_D)|\Omega_d|^2}, \tag{4}$$

and the common group velocity of E_a and E_b becomes $v_g \simeq |\Omega_d|^2/(\alpha_0\gamma) \ll c$. Since the Doppler width $k\overline{v}$ is assumed to be smaller than the detuning Δ_D, the averaging over the atomic thermal motion has practically no bearing on Eq. (4).

The real part of the complex polarizability α_j is responsible for the phase shift ϕ_j of the corresponding field, $\phi_j(z) \simeq \mathrm{Re}(\alpha_j)z$, while the probability of the absorption p_j of the field depends on the imaginary part of α_j, $p_j(z) \simeq 1 - \exp[-2\,\mathrm{Im}(\alpha_j)z]$. From Eq. (4) we see that if one of the fields propagates alone in the medium, its absorption and phase shift are zero [13]. By contrast, if both fields are present, each of them induces an absorption and a phase shift on the other. One can verify from Eqs. (4) that $\mathrm{Re}(\alpha)/\mathrm{Im}(\alpha) = \pm\Delta_D/\gamma$, i.e., for $\Delta_D \gg \gamma$, the *phase shift is the dominant process* and absorption can safely be neglected. This can be realized taking advantage of the common *anomalous Zeeman effect*, which corresponds to $g_L \neq g_U$ and thus $\Delta_D \neq 0$, as, e.g., in ^{23}Na or ^{87}Rb atoms. Then we obtain

$$\mathrm{Re}(\alpha_{a,b}) \simeq \pm \frac{2\alpha_0\gamma|\Omega_{b,a}|^2}{\Delta_D|\Omega_d|^2}, \tag{5a}$$

$$\mathrm{Im}(\alpha_{a,b}) \simeq \frac{2\alpha_0\gamma^2|\Omega_{b,a}|^2}{\Delta_D^2|\Omega_d|^2}. \tag{5b}$$

In the discussion pertaining to the collinear Doppler-free setup (Fig. 1, left inset), we have neglected the interaction of the E_a and E_b fields with the atom via the transitions $|1\rangle \to |5\rangle$ and $|3\rangle \to |5\rangle$, respectively, due to the large detunings $\pm(\Delta_L + \Delta_U)$. This simplification leads to the neglect of the self-phase modulation of the weak optical fields, given by

$$\mathrm{Re}(\alpha_{a,b}) \simeq \pm \frac{\alpha_0\gamma|\Omega_{a,b}|^2}{(\Delta_L + \Delta_U)|\Omega_d|^2}, \tag{6}$$

which is a weaker effect than the cross-phase modulation of Eq. (5a). Furthermore, this self-phase modulation does not depend on the presence of the other field, which allows us to separate it from the cross-phase modulation. For some applications, however, such as generation of optical solitons and phase conjugation [1], this self-phase modulation may be important and interesting in its own right. We stress that the self-phase modulation is absent in the setup of Fig. 1, right inset.

As a concrete example of cross-phase modulation, consider the vapor of ^{87}Rb atoms, where the pertinent lower and upper levels are $5S_{1/2}, F_L = 1$, and $5P_{1/2}, F_U = 1$, with gyromagnetic factors $g_L = -\frac{1}{2}$ and $g_U = -\frac{1}{6}$, respectively, and the transition frequency is $\omega_0 \simeq 2\pi \times 3.775 \times 10^{14}$ rad/s. Let us choose $N = 10^{14}$ cm^{-3}, $|\Omega_d| = 5 \times 10^6$ rad/s, and $\Delta_D = 70\gamma$, corresponding to $B \simeq 430$ G, $\Delta_L \simeq 2\pi \times 3 \times 10^8$ rad/s, and $\Delta_U \simeq 2\pi \times 10^8$ rad/s, which are smaller than the hyperfine splittings of the lower and upper atomic levels, 6.8×10^9 s^{-1} and 8.1×10^8 s^{-1}, respectively. Yet the Doppler width $k\overline{v}$ of the atomic transitions should be at least few times smaller than Δ_D. For the chosen magnetic field B, this sets the upper limit on the temperature of the atomic gas, which for the parameters listed above is $T \leq 10$ K. Stronger magnetic fields would allow for higher temperatures, at the expense of longer propagation distance or higher density. For the present values, two focused *single-photon beams* E_a and E_b (beam cross section $\sigma_{a,b} \simeq 10^{-8}$ cm^2) of a microsecond duration can induce a mutual phase shift of the order of π over a distance of ~ 3.8 cm, while their absorption probability remains close to zero, $p_j < 0.1$. In the setup with cold atomic gas (Fig. 1, right inset) we obtain the same phase shift π and absorption over a distance of propagation of ~ 1 cm [14], corresponding to the interaction of the fields with $\sim 10^6$ atoms.

Next we consider the cross-absorption scheme. The atomic level configuration is the same as in Fig. 1(b), but the frequencies of all the fields are lowered by Δ_D, i.e., $\omega_{d_1} = \omega_0$, $\omega_{d_2} = \omega_0 - 2\Delta_D$, $\omega_a = \omega_0 - \Delta_L + \delta_a$, and $\omega_b = \omega_0 - \Delta_L + 2\Delta_U + \delta_b$. Here again we have Raman resonances on the two-photon transitions $|2\rangle \to |1\rangle$ and $|2\rangle \to |3\rangle$. The character of the cross coupling is, however, different, since the E_a field is now resonant with the atomic transition $|3\rangle \to |5\rangle$, while the E_b field is detuned from the $|1\rangle \to |5\rangle$ resonance by the amount $2\Delta_D$. The Stark shifts of the states $|1\rangle$ and $|3\rangle$ due to the off-resonant interaction with the fields E_{d_2} and E_{d_1} are given by $|\Omega_d|^2/3\Delta_D$ and $-|\Omega_d|^2/\Delta_D$, respectively, which can be incorporated in the detunings $\delta_{a,b}$. Only the perpendicular arrangement of the $\sigma_\pm$ polarized weak fields and π polarized driving fields in a cold atomic gas (Fig. 1, right inset) is suitable for the cross-absorption scheme, since the frequency of the E_b field exactly matches the frequency of the atomic transition $|2\rangle \to |4\rangle$ which, in the case of collinear geometry, will induce a strong, resonant, unconditional absorption of that field. Equations (1)–(3) still apply upon making the indicated changes.

In the cross-absorption case, similar to the case of cross-phase modulation, we solve Eqs. (1) perturbatively in the weak-field limit $(A_2 \simeq 1)$. At the Raman resonance for both fields, $\delta_a = \delta_b = 0$, we then obtain for the imaginary part of the polarizabilities

$$\mathrm{Im}(\alpha_{a,b}) \simeq \frac{\alpha_0|\Omega_{b,a}|^2}{|\Omega_d|^2}. \tag{7}$$

In deriving Eq. (7), we have assumed that $\gamma/\Delta_D > |\Omega_{a,b}|^2/|\Omega_d|^2$, which is well satisfied for the parameters listed above. Thus, if only one of the fields propagates in the medium, its absorption is vanishingly small. By contrast, if both fields are present, each of them induces a *strong absorption* of the other. With the experimental parameters given above, the induced absorption depth of a single-photon pulse is $\sim 4.3 \times 10^{-3}$ cm, i.e., the fields' intensities are reduced by a factor of e after they have interacted with only 4300 atoms.

The treatment outlined here has focused on the essential aspects of our scheme, yet certain experimentally relevant issues have to be addressed briefly.

(a) Diffraction. Tightly focused Gaussian beams $E_{a,b}$ would normally diffract over the distance $\sigma_{a,b}\omega_0/2\pi c \simeq 1.3 \times 10^{-4}$ cm, which is much smaller than the propagation lengths necessary for achieving the desired phase shift and absorption. One can, however, take advantage of the long-distance diffraction-free propagation of weak Bessel beams. Alternatively, one can use the focusing properties of EIT.

(b) Adiabaticity. The adiabatic solution (4) of the amplitude equations (1) is justified by the fact that we have considered weak (much less that one photon per atom), slowly varying ($\tau \gg \gamma^{-1}$) fields $E_{a,b}$. In the case of intense and/or short-duration pulses, however, only the time-dependent treatment of the coupled set of Maxwell and density-matrix equations will rigorously solve the problem.

(c) Spectral broadening. Since the phase shift of each pulsed field is proportional to the intensity of the other, it will be maximal at the pulse peak and vanish at the tails. This will produce frequency chirping of the pulses and, therefore, their spectral broadening. One has to take care that the resulting spectral width does not exceed the transparency window of the EIT resonance $\sim |\Omega_d|^2/\gamma$.

(d) Entanglement. The two cross-coupled beams can become entangled, by splitting one of them into two components, one of which passes through the cell and interacts with the other (Fig. 1, left inset). It has been shown for the case of cross-phase modulation that a phase shift of π with negligible absorption should render two coherent or single-photon beams entangled [10]. In the case of cross absorption, our scheme (Fig. 1, right inset) can serve as a very sensitive conditional photon switch [7,11], whose sensitivity is limited by the free-space shot noise and the detector efficiency.

To sum up, we have shown that a simple scheme, comprised of a transverse static magnetic field and two optical driving fields, can create a new regime of *symmetric*, extremely efficient nonlinear interaction of two weak pulses in atomic vapor, owing to EIT via Zeeman-split levels. The resulting giantly enhanced cross-absorption and cross-phase modulation may open the road to the development of novel Kerr shutters and phase conjugators, as well as to quantum information applications [2] based on absorptive or dispersive two-field entanglement [7–11].

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[14] In the cold-atom setup with circularly polarized weak fields $E_{a,b}$, all the dipole-matrix elements of the $F_L = 1 \leftrightarrow F_U = 1$ transitions have the same absolute value $|\mu_{ij}| = \mu$, while in the collinear Doppler-free setup with linearly polarized weak fields, all the dipole-matrix elements of the $\Delta M = \pm 1$ transitions have the absolute value $\mu/\sqrt{2}$.